

Author's reply to referee comments given on Svensson et al. "Contribution of dust and elemental carbon to the reduction of snow albedo in the Indian Himalaya and the Finnish Arctic."

Here we present point-by-point responses to the referee's comments. The referee's comments are presented in plain text in this document, while our reply is indicated in italics.

The consequent changes made according to the referee comments have improved the revised manuscript. Deletions in the manuscript have been marked by ~~striketrough~~, whereas additional text are indicated by **red color**.

On behalf of my coauthors, yours sincerely,

Jonas Svensson

Anonymous Referee #1

The paper is focused on a significant and compelling issue: the attribution of light absorption in snow between primarily natural sources (mineral dust), and sources strongly influenced by anthropogenic activities (EC, especially in India). There are several main points made in the paper:

- 1) Mineral dust is perhaps the dominant light absorbing impurity in Himalayan snow
- 2) MAC of EC in snow is lower than for laboratory EC
- 3) MAC of EC decreases with increasing snow depth.

Although I imagine that points 1 and 3 have a good chance of being correct, all three of these conclusions suffer from some lack of support in the paper. I have substantial concerns about some of the assumptions and interpretations made in the paper, and question the conclusions that can be drawn from it.

My concerns are focused on the following issues:

- 1) The EC measurement of with the TOM method may be significantly affected by carbonates in the snow.

The authors cite Cavelli et al., 2010 to suggest that any contributions would show up as OC in the analysis, but this is not supported. First, Cavelli et al deals with atmospheric aerosol analysis in which carbonate “is generally <5% [of the aerosol]”. However in snow, and especially in the Himalaya, radically more carbonate can be present. For example see: Di Mauro, B., F. Fava, L. Ferrero, R. Garzonio, G. Baccolo, B. Delmonte, and R. Colombo (2015), Mineral dust impact on snow radiative properties in the European Alps combining ground, UAV, and satellite observations, *J. Geophys. Res. Atmos.*, 120, 6080–6097, doi:10.1002/2015JD023287. Which indicates that mineral dust content in snow in the European alps varied from the cleanest conditions (3 ppm dust) to levels of 200 ppm. 3 ppm of dust represents a factor of 15 times more mass than the EC concentration estimates of Svensson et al in Figure 12 (which I estimate as 200 µg/liter = 0.2 ppm EC). Note, too, that Cavelli et al points out for calcite that it “will possibly be detected as EC”, and pretreatment of ice core samples was a prerequisite for analysis in Ming, J., H. Cachier, C. Xiao, D. Qin, S. Kang, S. Hou, and J. Xu (2008), Black carbon record based on a shallow Himalayan ice core and its climatic implications, *Atmos. Chem. Phys.*, 8(5), 1343–1352, doi:10.5194/acp-8-1343-2008. As yet, the uncertainty raised by this issue is unbounded. Obviously, high mass ratios of mineral dust to EC in the Himalaya that contribute significantly to the TOM EC measurement are a real possibility that must be seriously addressed; ideally the authors would test some filter punches of ambient Himalayan samples with and without the type of acid treatment used in Ming et al., to constrain this issue. Note that conclusion #2 above is only valid if the EC determination is accurate. Further, note that conclusion 1, is not weakened by the carbonate issue, but should be reframed to reflect the clear contributions of mineral dust to in-snow absorption in the Himalaya.

The issue of carbonates in samples (both air and snow) is complex and further work on the topic is needed. During OCEC-analysis with the Sunset instrument and using the EUSAAR_2 protocol (used here) carbonates show up as a fourth peak during the first phase when OC is detected, and so it will affect OC (as pointed out by Cavalli et al., 2010). In other protocols (as well as other instruments), such as IMPROVE there is a risk that carbonates will affect the EC detection stage. It therefore depends on which protocol and thermal-optical instrument used (IMPROVE is frequently associated with DRI-analyzer). Using fumigation is a possibility, as in Ming et al. (2008), but in our case in favor of conserving resources we have not made the tests with acids. Instead, we have inspected the thermograms from OCEC-analysis, by visually observing any fourth peak in the

first stage, and have not observed any fourth peak with significant contribution to thermogram area for our samples. Furthermore, a recent study by Zhang et al. (2017), actually made tests as outlined by the referee here. With a subset of their filter samples from Tibetan snow, Zhang and colleagues found little difference between samples measured with and without acidification (less than 20% discrepancy).

In our revised manuscript we have added this reference, and clarified the text that carbonates are assumed not to be present insignificant amounts in our samples.

2) The laboratory tests of EC and SiC do not strongly constrain uncertainties in evaluation of ambient snow samples. The authors present several tests to evaluate the performance of their three measurement types: OCEC-analyzer TOM determination EC concentration; PSAP measurement of loaded-filter light transmission before and after undergoing TOM analysis; and loaded-filter light transmission measured with the OCEC-analyzer. However, it is not clear how relevant these tests are to the conditions relevant to the ambient samples. Comparisons of optical depth measured with the PSAP and the OCEC were presented as if they indicate the validity of the EC determination from the optical measurement; in fact they only illustrate measurement precision since both instruments were quantifying the same thing (optical depth on the filter). Hence the statement “The good agreement between the two optically derived EC values suggests that much of the scatter seen in Figure 5 is due to the uncertainty in the analyzed content of EC using TOM (and FID)” is incorrect and misleading. The authors should re-analyze the optical/TOM EC comparison to attempt to ascertain the confounding influences of the SiC, even as an imperfect proxy for ambient mineral dust contaminants. Further, although the EC concentration was not known (page 10, line 5), the relative concentration was. Hence there should be better constraints on the influences of the SiC on the different determinations of EC loading; I expect this dimension of analysis also to be relevant to the ambient samples. Undercatch by the filter was mentioned, but not given sufficient attention to justify confidence in the dataset. This can be a major problem (close to 100% for some filters!), especially for BC which is largely in the $<1 \mu\text{m}$ mode, and typically $<0.5 \mu\text{m}$; hence the statement in the paper “smaller particles normally contribute little to total particulate mass” is not relevant to BC and filter undercatch. Note that the linearity of optical depth with estimated EC loading does not constrain filter losses. At a minimum, the authors should test undercatch with their filters by re-filtering post-filter liquid from one of their EC standards with a high-efficiency filter. See, for example, Schmitt et al., doi:10.5194/tc-9-331-2015 for a treatment of similar issues.

The laboratory experiments are highly relevant for the ambient conditions, although the exact properties of the mixture of aerosol on the ambient filters are not known. It is imperative for the interpretation of field measurements that we can demonstrate consistency in the method using laboratory conditions that are controlled. Every permutation of possible aerosol mixtures can of course not be tested and we chose a simple set up where BC and the mineral proxy have roughly the type of optical properties that can be expected in ambient samples. Without this step, interpretation of snow samples associated with many unknowns would be very difficult. It is true that a best guess MAC has to be used for the estimation of light absorbing dust mass concentration but the variability and thus accuracy of using a fixed MAC would be the focus of another study.

The statement that the “good agreement” in fig.5 between the two optical measurements indicate that most of the scatter between TOM and PSAP is related to where the OCEC instrument determines to place the split point between OC and EC, still holds. The reviewer is right in that we do not explicitly suggest the reason for this scatter. Undoubtedly, the fact that BC is mixed with a mineral could very well influence the results. This is also one of the underlying reasons for why the laboratory exercise presented in the study is so central. We clarify the statement by noting that the reason for the scatter could be related to the fact that BC is mixed with SiC.

Undercatchment by filters is a potential uncertainty with loss of small particles. Many studies (e.g. Lavanchy et al., 1999; Forsström et al., 2013; Lim et al., 2014; Torres et al., 2014; Kuchiki et al., 2015) have performed limited tests on this enduring issue, and an array of different results have been presented, with nearly 100% inefficiency or efficiency to capture the particles on filters. Clearly, the issue is complex, depending on several factors and still remains unresolved, and the aim of this paper is not to solve it. For our work here we are confident that larger sized particles have been captured by the filter, whereas we are unsure how efficiently our filters have collected smaller sized (<100 nm) particles. In the revised manuscript the reviewers concern has been elaborated on further.

3) Conclusion 3 above is based on the results shown in Figure 13. In my opinion, the data are not statistically robust enough to support that conclusion, and the discussion in p. 23 lines 28-34. Due to these major issues, I believe that the abstract, discussion, and conclusions should be modified dramatically.

We are aware that this is based on one pit study only, which is not enough and a lot more work needs to be done to obtain better statistics. However, with our data, and the one snow pit, we demonstrate a possible tendency (and not trend with the data). In the revised manuscript our enthusiasm of the results have been softened, highlighting the need for more measurements and that our tendency is speculative.

Smaller comments:

Abstract: 1) Snow albedo is strongly dependent on many factors beyond LAI (eg snow morphology, snow depth, underlying surface properties); since albedo is not a focus here, I suggest that the first sentences be reformed.

It is true that snow albedo depends on many factors. Still, LAI have the potential to significantly affect the albedo, depending on the pre-existing circumstances. As referee #3 also suggested, this sentence is changed in the revised manuscript.

Introduction 1) Since the SP2 is not used here, including it at this depth in the introduction seems unnecessary.

Actually, we disagree with the reviewer. This paper have in it a significant technical aspect of understanding LAI in snow and it is therefore pertinent to describe other relevant methods with a sentence or two. We adhere to the referee's criticism by shortening this particular section in the revised manuscript.

2) The discussion of non-BC LAI (page 4, lines 21-35) should probably be introduced before discussing the measurement techniques.

That is a good point, thanks. This section has been moved in the revised manuscript, appearing before the measurement methods of LAI section.

Misc:

1) Page 6 line 1: What is meant by "protected" here?

A glass container with a lid was used. We changed the word "protected" to "enclosed" in the revised version.

2) How were the filters dried?

The filters were dried in ambient conditions inside petri dishes. This has been added to the text.

3) P. 7, line 6: I am surprised that it is difficult to gravimetrically determine the dust concentration, as dust is almost certainly the dominant mass source left on the filter (at least in the Himalaya, where the optical thickness was too large to be measured!).

It is true that dust can be the dominating mass of the impurities on the filters. From our tests weighing filters show that these filters are not suitable for this procedure. Too much variance (noise) is introduced to get a good signal to noise ration. Additionally, the quartz fiber filters used are very fragile, and so while filtering, mass can easily be lost.

4) More information about the SiC used is needed. Manufacturer? Color? Etc.

The manufacturer is Carborundum (company non-existent anymore), as already indicated and the color was light grey with hints of blue. In the revised we added the color.

5) Please specify in the figure axis labels the technique (e.g. "optical" or "TOM" used to extract the various quantifications. For example, on figure 3, the caption merely indicates the EC was from the OCEC instrument, but it was not immediately clear that this via TOM.

Thanks, it is now fixed.

6) Page 12 line 7: again, the atmospheric results are likely not relevant in the Himalaya. I think they can be cited only for the Arctic results.

Our intent was to state that our laboratory measurements which showed that SiC has 100 times less MAC than BC, agrees with previous reports from atmospheric measurements (on a general scale), where dust is less absorptive.

7) What is the author's estimate of the loading of mineral dust on the ambient filters? How does this compare to the range tested in the laboratory?

We can only compare them by their optical signal. We can say nothing about mass. To do the latter we need to know the optical properties of dust in India (or Finland for the other samples). Since we do not know, we have used the ratio of tau to say how much the mineral contribute. Our range in the laboratory probably cover the same range, but this is speculative.

8) Please provide estimates of statistical relevance of the fits (e.g. r^2 values).

In the graphs the r^2 values are already presented.

9) I saw concentrations presented as $\mu\text{g/L}$ as well as ng/g . Please harmonize to a single unit for the reader (obviously these are effectively the same. . .)

Good, this should be consistent and has now been changed.

10) Page 17: lines 12-18: this is a lot of speculation without much reason for it. Suffice it to say that your results, for a different area where high spatial variability can be expected, are higher than a previous estimate.

Yes, that is true. But still we wanted to highlight some of these potential aspects that affect the differences. In the revised manuscript the text is changed accordingly.

10b) Page 17 lines 19-26: please include broader arctic estimates of EC concentrations for context. E.g. Doherty et al., 2010 ACP, doi:10.5194/acp-10-11647-2010.

Since Doherty et al. use a different measurement method we think it is sufficient to put our obtained results into a greater context with the results from other studies using the same measurement techniques. The other cited works present results for the broader European Arctic, which was not noted previously in the manuscript. This is now emphasized in the revised manuscript.

11) I don't see the value of the comments of page 17 lines 27-33, or figure 8 at this point in the text. This appears merely a technical point about measurement precision (as discussed above).

We disagree with the referee. The text is needed here to provide further credibility in our approach to analyze the data in the manner that we have chosen to do, so we feel that they are necessary to be there.

12) Couldn't the differences discussed in page 18 line 4-17 also be due to EC overestimation for example due to carbonate impacts?

As pointed out above, carbonates will not influence the TOM EC estimate, but could cause overestimation of OC. We refer to the discussion above for this point (p. 2-3 in this document).

13) Schwarz et al. only presented a theoretical estimate of MAC, not any measurements. Hence this is only suggestive, but it is also possible that the filter selectively catches larger BC. Please consider removing figure 10, as this is quite peripheral.

That is correct about Schwarz and colleagues, and in the revised manuscript this has been changed. The set-up was the same for the laboratory filters and the ambient filters, therefore the bias towards larger sized BC particles would exist in both sets of filters.

14) Page 20, line 18: "modes" is clearer than "peaking fractions".

That is true, and it is changed in revised manuscript.

15) Please specify the wavelength of the optical depth measurement in the OCEC instrument. How does this compare to the 532 of the PSAP? In discussion of relative absorption of mineral dust and BC, it is important to continue to specify the wavelength range for which the discussion is relevant.

For the OCEC-analyzer the laser operates at 632 nm (stated on page 7 in manuscript). Most of the discussion is for the PSAP, which is at 526 nm. This has been added to the dust fraction section (3.2.2) in the revised manuscript.

16) Please be consistent in referring to the dust "absorption" fraction. For example, in the caption to figure 11, "dust fraction" could be misunderstood by an un-alert reader as a mass fraction.

Referee #3 commented on this as well. The caption has been changed.

17) Is it possible to add data points to figure 12 a, so that the spread in results will be obvious to the reader?

Yes, an updated figure including the data points is now included.

18) What is the explanation for the huge differences in EC via TOM and OPTICAL at the sub surface contaminant layer shown in figure 12B?

This comes back to the OCEC-analyzer and where it places its split point in EC TOM, which has been shown to be less accurate for filters with a higher aerosol loading (Cavalli et al., 2010).

Anonymous Referee #3

Svensson et al. use a combination of a thermal-optical detection method and a particle soot absorption photometer to determine elemental carbon mass, its mass absorption cross section, and the contribution of mineral dust to the optical thickness of light absorbing impurities in snow. They conducted a series of laboratory assays with chimney and standard soot, as well as two different types of mineral dust, and mixtures of the various components to test the instrumental set-up. Subsequently, the method was applied to ambient snow samples from the Finnish Arctic and two Indian glaciers. The main findings are: a) the MAC of EC in snow seems to be lower than that of the laboratory test soot; b) dust plays a larger role in light absorption on the Himalayan glaciers than in the Arctic; and c) the MAC of EC in snow seems to increase with deposition age.

Generally, the study follows a careful design and is well described. It addresses the very important challenge of how to quantify the albedo-reducing effect of elemental carbon and mineral dust and their combination in snow, and contributes to the understanding of uncertainties by highlighting the importance of accurately determining the EC MAC in snow. Nevertheless, the work has some shortcomings: It is not clear why the MACs of two very specific dust types are determined in the laboratory and discussed when this is meaningless for the ambient dust samples with unknown absorption characteristics. An explanation of why the MAC of EC in snow is variable is missing, even though this is featured as a main result. Also the observation of a potential trend in the EC MAC value with snow pit depth is not backed up sufficiently. There is only a small number of data points, only one snow pit is discussed and the statistical significance of the trend is not given. Further, the value of addressing Arctic and Himalayan snow samples together in this manuscript is not evident. Additionally, the manuscript language needs to be improved.

I recommend addressing the above and following comments before a publication can be considered.

General comments

The title implies a discussion on snow albedo reduction through EC and mineral dust. However, the manuscript does not provide actual values of snow albedo reduction but rather focuses on accurately determining the mass of EC in snow and the contributions to optical thickness of EC and mineral dust. I suggest changing the title accordingly.

Title has been changed.

p. 10, l. 16/22: Information on light absorbing constituents in SiC and stone crush are needed. The light absorption of mineral dust is strongly influenced by e.g., hematite, goethite etc. There is no point in determining a MAC of a "random" dust sample in the laboratory to apply it to the ambient samples with unknown contents of light absorbing constituents as the authors state themselves on p. 20, l. 13. It is not clear, why dust samples are tested in the laboratory that have no relevance for the ambient dust samples. Also, in Figure 7, the discrepancy between the gravimetrically determined SiC mass on filters and the estimated based on the MAC when values are $> 7 \text{ g/m}^2$ are just discarded without any further discussion. Is it possible, that the method does not work for high dust loadings? I suggest that the dust related aspects of the laboratory assays are drastically shortened to the information relevant for the ambient samples.

As previously stated to referee #1 the laboratory experiments are highly relevant for our work and are needed to demonstrate that our measurement method works in laboratory conditions before analyzing field samples. Please see our response to referee #1 on p. 3 where this is elaborated on. For higher dust loadings the method is less accurate, as expected, since there is a lower signal to noise ratio for the optical measurement.

l. 13 – 27: What about local dust sources? There are most likely bare rock or mountain walls from which mineral dust can be deposited on the glacier. In general dust sources and in particular local dust sources can be highly variable so that any kind of interpretation in terms of trends is difficult, especially if no information about the origin is available. If the authors had mineral dust size distributions at least a statement on potential role of local sources if very large particles are present could be made.

True, this sort of statement(s) could be made if we had such data, but that is not the case.

l. 28 – 34: The interpretation that the MAC is decreasing towards the top of the snow pit is not convincing and an explanation why this might be the case is missing completely. There are not enough data points to conclude a trend in the MAC. The profile rather shows that LAI deposition is highly variable. Towards the bottom of the snow pit the ratio is also lower and if the point at 10 cm depth were not as low as observed, probably no trend would be inferred. The authors introduced on p. 20 the hypothesis that potentially large loadings on filters play a role for the observed variability of the MAC (see comment further above). To test this and to develop a potential explanation why the MAC changes, I recommend plotting in Figure 13 the ratio of the optical and TOM EC versus the ratio of Dust/EC and OC/EC to check if there is a relation with the overall LAI content. Also, indicate in the figure which ratio is meant. In the conclusion, the potential reasons for the observation should be given as well.

We realize that this is based on one pit and that our data is not conclusive. Referee #1 also commented on this topic (p. 4 in this document), further details are presented there.

The authors recommend additional work to constrain the optical properties of EC in snow but do not say how this could be done. Some more precise ideas would be useful.

That is true. In the revised manuscript we have added a thought on this.

Specific comments

p. 2, l. 3: What do you mean with “impact on climate”? LAI deposited on snow do not directly impact the climate but rather the hydrological cycle. Only if seasonal snow and glacier retreat significantly, local climate will change and LAI are hardly the major cause for it.

We did not mean it the way the referee has interpreted the sentence. LAI will affect climate through its lowering of albedo and subsequent snowmelt. Nevertheless, the sentence is now changed in the abstract.

p. 3, l. 2-3: Specify which radiation budget you mean and through which mechanism snow melt can be enhanced?

Snow melt is enhanced via snow darkening that is induced by the deposition of LAI. This has been clarified in the revised text.

l. 6: “In this context” refers to mountainous glaciers. Why are mountain glaciers more important than seasonal snow or ice caps?

We do not mean to suggest that mountain glaciers are more important than seasonal snow or ice caps in any way, and the text “In this context” has been removed in the revised version.

l. 12-14: I suggest removing this sentence and focusing only on BC effects in snow. In addition, BC has health effects everywhere where a human being is exposed, not only in cities and where open cookstoves are used.

This is true, the sentence is now deleted.

l. 21: is Bond et al. 2007 really the most up to date reference?

Bond et al., 2013 is newer reference sufficient for this statement.

l. 34: more elaboration on the thermal-optical analysis is needed here.

At that stage in the manuscript we simply want to only introduce the method. An elaboration is provided in the methods section, which we feel is adequate.

p. 4, l. 3 – 8: The description of the SP2 can be shortened because this instrument does not play any role in the set-up of this work.

In the revised version it has been shortened (as pointed out by the other referee, p. 4 of this document).

l. 10f: An explanation of why it is important to keep the samples frozen before analysis is needed.

Good point. Particle losses can be very significant if a sample is not kept frozen. In the revised manuscript we have added this.

l. 18f: The statement that small particles contribute little to particle mass in this context might be a bit misleading. More elaboration is needed. What is meant by small particles, what is the threshold that Lim et al. (2014) refer to? If it is < 100 nm the statement is ok, if it is < 300 for example, it would not be, because of the size distribution of BC. Also the last sentence does not take into account that the TOM method will also quantify OC, which can also absorb light and reduce snow albedo.

We are referring to fig. 3 of Lim et al. (2014), in which one can see how the filter efficiency increases with BC size. The issue of undercatch can be significant for quartz-filters and it is not resolved to-date. In the revised manuscript this has been modified.

p. 5, l. 21: atmospheric concentrations of what?

It refers to BC concentrations and other aerosol particles (particulate matter), which is stated earlier in the sentence, with both displaying seasonal patterns.

p. 6, l. 3f: A description of how the filters were dried is missing here and later on for the microwave heated samples as well.

As noted by the other referee also, this has now been modified in the revised manuscript.

Figure 1: The location of the glaciers and the valley is not really visible, the site symbols are too small and the lat/lon numbers in the middle of the plot are distracting. Also the location of the sampling sites in Finland are not visible at all. Think about including two maps that show the details for each region, while keeping the global map.

A new map has been made for the revised manuscript.

p. 7, l. 6: Why is it difficult to gravimetrically determine the dust load on filters? This is a standard method and the mass of dust very often is much higher than the mass of other LAI. Do you mean it is difficult in regions where little dust is present?

From our tests it is difficult to weigh the quartz-filters as there is significant variance. This issue was also commented on by referee #1, so please see our response on p. 5.

l. 33: It is unclear from the description what the original value of transmittance is.

It refers to the transmittance signal measured by the OCEC-analyzer right before the first step of analysis (OC detection). We have added the text “(measured before thermal sequence starts)” in the revised manuscript to clarify this.

p. 8, l. 5ff: Why are the authors confident that no acid treatment of the samples is necessary to eliminate carbonates? In ambient mineral dust calcium carbonate or other carbonates are a common constituent which might strongly affect the OC/EC analysis and not only the quantification of OC as you explain in l. 11f (split point).

Carbonates shows up as fourth peak during the first stage (OC-stage) of OCEC-analysis using the EUSAAR_2 protocol (Cavalli et al., 2010). This was not found for our samples after visually inspecting our thermograms from the analysis. See also comment by referee #1 and our response on page 2 for more information.

l. 8-12: Do the authors have any estimate of the loss of small particles at least for the laboratory experiments using the same filters? Since the laboratory pre-assays are meant to quality control the results of the ambient samples this is a crucial piece of information that should be obtained.

We do not have any estimate of the loss of small particles. This is a topic (undercatchment) that should be further explored, but not in this paper.

l. 18 – 21: From the description the methodology is not clear. Did the authors use duplicate samples, so 2 times original and 2 times heated, or two punches and one was heated while the other wasn't?

This is the general methodology of the PSAP and how it was applied for our purpose. It is further described on p. 10 in the manuscript. In the revised manuscript we have directed the reader that the methodology is further presented in section 2.3.

p. 9, l. 9f: What is the reason for this? Is the effect of the filter so overwhelming that the scattering effect of the actual sample is by far outweighed?

The reason is that the filters we are using do not have any known correction factors (compared to for example PSAP-filters that have existing correction factors in the literature). As we note in the previous sentence of the manuscript, we treat the enhancement factor as a constant for our samples, and so it is included in our effective MAC.

l. 20: It needs to be stated more clearly that the MAC values reported here cannot be compared to other studies.

This is now emphasized in the revised manuscript.

p. 10, l. 8: Information on the homogeneity of the chimney soot is missing. How comparable are samples? Why was chimney soot chosen, is it representative for soot deposited on snow in Finland and India?

Samples are comparable in the sense that when a larger amount of the stock solution was taken, the filter appeared darker (and had higher EC content). Chimney soot was chosen since it is easily accessible and is a

common pollutant in the study area, especially in the Himalayas. Moreover, if a mixture of chimney soot, vehicular soot and other soot, then what would be the appropriate proportions?

L. 35 f: What are twin samples, and what does “separate instruments” refer to? The meaning of the sentence is unclear.

The “separate instruments” refers to the OCEC-analyzer and the PSAP, and the twin samples refers to two identical samples. In the revised manuscript this has been clarified.

p. 11, l. 1: how many filters were tested and what was the result in numbers?

The number of filters was 7 with a difference less than 5%.

l. 19: What is divided by Cref?

It is the optical depth as noted later in the sentence.

Figure 4: How would the slope look like if the point with the highest loading of SiC were not considered? It drives the correlation result and how would results look like that are presented in Figure 7?. In the caption the wavelength information is missing.

Excluding the point with the highest SiC does not change the slope (it remains 0.23). The wavelength information is now included.

Figure 5: Add the 1:1 line.

In the revised manuscript this has been added to the figure.

p. 14, l. 8ff: The spread in the comparison of the two optical EC determination methods is not necessarily an indication for variability in the evolved carbon determination of EC. It is not clear what is meant with “uncertainty in the analyzed content of EC using TOM. . .”. Which factors introduce uncertainty in this method? Which introduce uncertainty in the optical methods?

A similar comment was noted by referee #1. We are referring to that that most of the scatter between TOM and PSAP is related to where the OCEC instrument determines to place the split point between OC and EC. See further our comment to referee #1 on page 3.

p. 17, l. 11f: Concentrations are given in $\mu\text{g} / \text{L}$ and ng/g , one consistent unit should be chosen.

Thanks, it should now be consistent in the revised manuscript.

Figure 8: Is the outlier at Sunderhunga taken into account for the linear regression? If so, what would the slope look like without? It would be even greater than 19 % and that is a relatively large deviation between the optical measurement of EC with TOM and the estimated EC based on the MAC and the PSAP data. What does this discrepancy mean for the validity of the methods?

The outlier from Sunderdhunga is included in the regression. Taking this data point out from the regression we obtain a slope of 1.31 (although we do not see the point of removing this one data point since it merely shows some scatter in the data). It could be argued that data points with an EC content higher than 0.03 g m^{-2} (ca. $\text{Tau}=1$) could be removed since at higher EC content there is a lower signal to noise ratio for the PSAP measurement. With this procedure we get an identical slope (1.18) as the original value in fig. 8 (1.19). Thus, removing data points does not affect the regression.

p. 18, l. 12: A reference and an explanation is missing.

This is the enhanced absorption effect of BC particles dependent on the mixing state. References have been added in the revised manuscript, and the sentence has been slightly modified for clarity.

p. 20, l. 9ff: The underlying assumption why EC is less absorbing in this case is not provided and not evident. Do the authors assume that the EC is embedded so thickly in OC that light does not penetrate to the EC? If the authors imply matrix effects of the sample, the question is how relevant this is to the ambient snow? On the filter the particle mass is concentrated and the packing of the particles on the filter is not representative for the packing in the snow.

We do not know the reason for the apparent reduction in absorptivity between our lab BC and ambient samples, this is why we offer different hypothesis for this difference. One of these is embedding BC particles in water-insoluble OC, the other is size effects.

It is most likely that the conditions on the filter differ from the conditions when the particles are naturally in the snow. However, we are not aware of any non-intrusive method to date that can determine LAI concentrations in snow and ice. In our study we never exploit our numbers into calculating radiative effects or climate impacts, but rather try to understand levels of LAI that can be present in snow and ice and advance our knowledge about possible processes related to this.

p. 21, l. 10: Do the authors mean the average EC concentration when saying “composite”? Be more clear.

Yes, that is what was meant, and is now changed in the revised manuscript.

p. 23, l. 3: How do the authors know that those are different seasons over various years and not melting and freezing cycles within one year? An explanation is missing in the text.

Concentration cycles as observed reflect seasons, while short-term events would produce more random distributions. In the previous sentence we offer an explanation.

l. 7: Water soluble constituents might percolate, this is not known because it hasn't been investigated. So there cannot be a statement that this is not the case.

The vertical LAI distributions indicate that not much mixing has taken place. We have changed the wording of the sentence and added a reference where this is discussed (Doherty et al., 2013).

Technical comments

p. 2, l. 3: write “with subsequent implications for snow melt. To more accurately quantify changes in snow albedo, . . .”

Changed.

l. 5-6: write “. . .from the Indian Himalaya and pared the results to snow samples. . .”

Changed.

l. 18: “dust deposition”

Changed.

l. 22: Do you mean deeper pits?

Yes, that was meant, and is now changed.

p. 3, l. 5: There is a newer AMAP report from 2015.

Yes, that is true, but that reference does not cover this topic.

l. 9: you should list rather fuel types than activities if you refer to carbon-based fuels.

We do not see the point in listing different fuel types. In the revised manuscript we added “in activities” to the sentence to clarify.

p. 4, l. 21: “can” instead of “may”.

Changed.

l. 22: “microorganisms” instead of “microbiology”

Changed.

l. 33: “Other methods consist of using transmitting light. . .”

Changed.

p. 5, l. 1: “in the Indian”

Changed.

l. 4: tests

Changed.

l. 8: “2.1.1 The Indian Himalaya”

Changed.

l. 12: “valley-type glaciers in the Ganges basin. . .”

Changed.

l. 14: delete “residing”

Changed.

l. 27: “Dust from local sources has. . .”

Changed.

l. 31: delete “designated”

Changed.

p. 6, l. 14: “sampled snow”

Changed.

l. 16: “(the first. . .”

Changed.

l. 17: “. . . where details of the area. . .”

Changed.

p. 7, l. 31: “ for pyrolysis (darkening of the filter) occurring during the. . .”

Changed.

l. 34: delete “filters” before EC

Changed.

p. 8, l. 14: “uses”

Changed.

l. 32: write micrometer instead of millimeter

Changed.

p. 10, l. 2: Start the sentence with “A series of . . .”

Changed.

l. 5: replace “minute” by “small”

Changed.

p. 11, l. 15: “dependent on”

Changed.

l. 16: delete “thus many influences on it.”

Changed.

l. 20: delete “somewhere”

Changed.

l. 21: “for our BC solution data”

Changed.

l. 22 “Bond et al. (2013) report. . .”

Changed.

Figure 3 Caption: “comparison of. . .”, here and in several other captions. Also, sometimes OCEC-analyzer, TOM or Sunset analyzer is used. A more consistent use of the method name is needed.

Changed.

p. 12, l. 4: “Figure 4 shows results analogous to Figure 3. . .”

Changed.

p. 13, l. 9: "The data are scattered,. . . regression is within 17 % of the 1:1 line."

Changed.

p. 14, l. 7: "As observed, the EC amounts derived by two optically different methods show a consistent relation. . ."

Changed.

Figure 6 caption: "between the optical measurement of EC. . . on the substrate using PSAP data and the . . ."

Changed.

p. 15, l. 9: "Two slopes are presented, . . ."

Changed.

Figure 7 caption: "containing all data points"

Changed.

p. 16, l. 14: "with material and quantitative impurity. . ."

Changed.

p. 17, l. 2: "these surface samples contained LAI mostly originating from the post. . ."

Changed.

l. 3: "studies of BC"

Changed.

p. 17, l. 19: "For reference, the EC concentration in the surface. . ."

Changed.

l. 22ff: ". . . in Pallas might result from the fact that the majority of samples was taken later in the snow season. . . and EC has likely concentrated. . ."

Changed.

l. 25: "On a larger scale, Northern Europe and Arctic, the concentrations. . ."

Changed.

p. 18, l. 9: "a smaller absorption efficiency", replace "absorbing efficiency" by "absorption efficiency" here and in several other places in the text.

Changed.

l. 17: delete "originating"

Changed.

p. 19, l. 9: “demonstrates”

Changed.

l. 12: “for BC we use the same complex. . .”

Changed.

Figure 10: Explanations for the symbols in the equation are missing.

Changed.

p. 20, l. 4: “liquid”

Changed.

l. 7: “a scattering medium shows enhanced. . .”

Changed.

l. 8: “was” instead of “were”

Changed.

l. 13: exchange “applicable” by “possible”

Changed.

l. 18: “modes” instead of “fractions”

Changed.

l. 19: “with modes at 35 % and 65 %.

Changed.

l. 20: “by LAI other than BC. . .”

Changed.

p. 21, l. 3f: “reach as much as 56 %.. in the Tibetan. . . as a fraction of the optical depth of LAI on the filter,. . .”

Changed.

l. 5: delete “an” before albedo.

Changed.

Figure 11 caption: “Frequency of the occurrence of dust optical thickness fractions at the three sampling sites.”
Otherwise the mass fraction might be inferred.

Changed.

p. 23, l. 2: delete “evidently”, replace “core” by “pit”

Changed.

l. 3: is “alternating” meant by “altering”?

Changed.

l. 8: replace “that” by “who”

Changed.

l. 13: replace “at best ca.” by “potentially”

Changed.

l. 30: “decrease” instead of “be decreasing”.

Changed.

p. 24, l. 14: “EC deposited on snow”. The EC does not originate from the snow.

Changed.

l. 14 f: “Our finding of a MAC value of about half of . . . EC particles, can have implications for the snow. . .”

Changed.

References

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